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Resonant exciton-phonon spectra in ZnCdSe/ZnSe single QW: Raman scattering and hot luminescence; extended and localized excitons

V. V. Travnikov[†], V. H. Kaibyshev[†], M. Rabe[‡] and F. Henneberger[‡]

[†] Ioffe Physico-Technical Institute, St Petersburg, Russia

[‡] Humboldt University, Berlin, Germany

Introduction

Exciton–phonon spectra at resonant photoexcitation of ZnCdSe layers have been investigated in a number of works [1, 2, 3] earlier. However, the interpretation of their formation mechanisms had rather conflicting character. Part of authors considered that the observed lines go through Raman scattering mechanism and the others treated them as luminescence lines. The nature of exciton and phonon states which are involved in the process of REP formation have been treated differently also.

In the presented paper we have investigated in detail fine structure and intensity of resonant exciton–phonon (REP) spectra in ZnCdSe/ZnSe QW structures as function of tunable laser energy at 8 K. We have found that in REP spectra two types of lines are presented simultaneously. The most intensive REP component correspond to Raman scattering through extended exciton states. Other less intensive REP components correspond to hot luminescence of localized excitons. Analysis of phonon modes which can be involved in formation of REP spectra in our strain QW structures has been carried out.

Experimental

The investigated samples consisted from single 5 nm ZnCdSe quantum well (Cd mole fraction $\sim 13\%$) which was sandwiched between 20 nm (cap) and 25 nm (buffer) ZnSe layers. Emission spectra have been measured at $T \sim 8$ K at photo-excitation by a tunable dye laser. Typical resonant emission spectra, taken on one of the samples, for different excitation energies E_{ex} in the energy region of the ZnCdSe ground (heavy-hole) exciton resonance (the position of this resonance is shown by the arrow E_{1hh}) state are shown in Fig. 1. The most characteristic feature of these spectra is the presence of strong lines at the energy positions $E_{emi} = E_{ex} - \omega_i$, where ω_i is the characteristic phonon energy. Besides these resonant exciton–phonon (REP) lines a rather intensive photoluminescence (PL) band corresponding to the luminescence of ground exciton state E_{1hh} is observed (see spectrum 3 in Fig. 1).

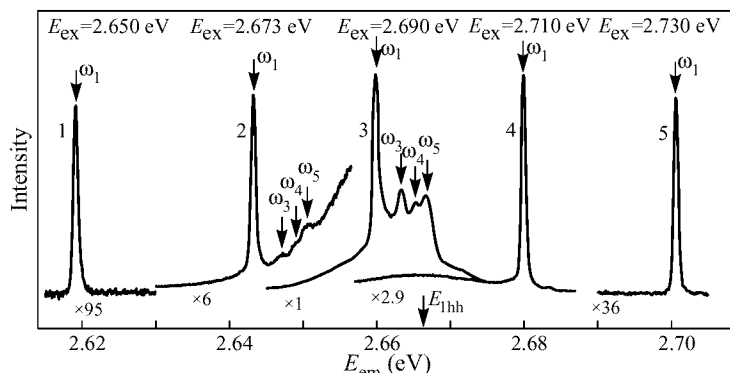


Fig. 1. Emission spectra of the QW sample at different excitation energies ($T = 8$ K).

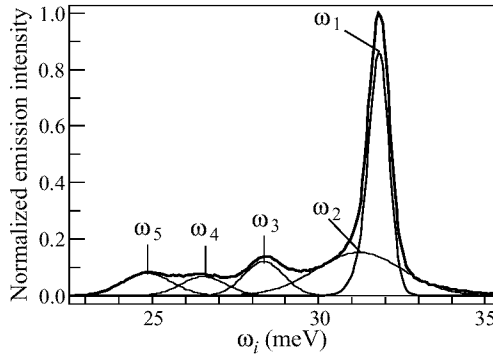


Fig. 2. Decomposition of one of the observed spectra.

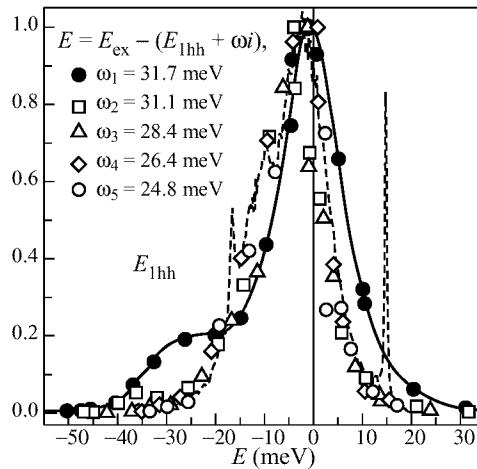


Fig. 3. Resonance profiles (filled and open circles) of the REP components. The dashed line is the PL spectrum. The solid line is a guide for eye only.

The intensity of the ω_3 – ω_5 REP lines correlates with the intensity of the PL background under these lines. The lines are observed only in the energy regions where the PL background has noticeable intensity (spectra 2, 3, 4 in Fig. 1). At excitation far from the resonance (spectra 1, 5) where the PL background intensity is negligible there are only the ω_1 lines in the observed spectra. By the subtraction of the PL background from a spectrum we have obtained spectra which correspond to “pure” resonant exciton phonon optical transitions. After that we have decomposed with high accuracy these spectra into five components (Fig. 2) and have measured the resonance profiles (emission intensity versus laser energy) for all obtained REP components. The resonance profiles of all components are characterized by pronounced outgoing resonance (resonance enhancement for emitted (scattered) light). As far as the energy of participating phonons are different the energy position of maxima of the resonance profiles are shifted relatively each other. Therefore to have possibility to compare the character of the obtained resonance profiles we have presented in Fig. 3 the intensities of the REP components as a function of the energy $E = E_{emi} - E_{1hh} = E_{ex} - (\omega_i + E_{1hh})$ (where $E_{1hh} = 2.667$ eV). Figure 3 clearly shows, that the resonance profiles for the ω_2 – ω_5 components (open signs) in the limits of experimental accuracy are practically the same but they are quite different from the ω_1 resonance profile (solid circles).

The horizontal axis in Fig. 3 corresponds to the energy of final (emitting) photon states relative to the resonant energy E_{1hh} . Luminescence spectra also can be presented as a function of the energy E_{em} of emitting states relative to the energy E_{1hh} ($E = E_{em} - E_{1hh}$). As a result we have possibility to compare the efficiency of the REP processes with efficiency of luminescence for the terminal (final) states with the same energy. Dashed line in Fig. 3 corresponds to luminescence intensity as a function of the energy $E = E_{em} - E_{1hh}$ for the PL spectrum which was obtained at the excitation energy 2.712 eV (sharp lines in the spectrum correspond to Raman scattering).

Besides the emission spectra we have investigated the excitation spectra of the investigated emission. Properties of these spectra also have been used for suggested interpretation.

Analysis and discussion

The resonance profile for the ω_1 component has characteristic shape with the pronounced outgoing resonance ($E_{ex} = E_{1hh} + h1$) and the incoming resonance ($E_{ex} = E_{1hh}$) as a shoulder. Such shape is a characteristic for resonant Raman scattering through free exciton states with simultaneous participation of elastic scattering on defects or impurities [4, 5, 6] and it was observed earlier at investigation of RRS in bulk semiconductors [4, 5] and in QW structures [6]. If the Raman scattering would involve only phonons alone the incoming and the outgoing resonance would have the same intensity [6] in contrast to our results. In our case the outgoing resonance is the dominant feature of the ω_1 resonance profile. It indicates to an participation of additional elastic scattering events in the Raman process [6]. In QW structures such elastic process can be connected with scattering on roughness [6]. At excitation above the exciton resonance the elastic processes lead to the scattering through the real exciton states. Participation of real exciton states with large density of states increases essentially the intensity of resonant Raman scattering [6, 7] and it is this that leads to the dominant outgoing resonance. Taking into account the shape of the ω_1 resonance profile and the fact that the ω_1 line is observed in all parts of the investigated energy region we suppose that in our case the ω_1 line correspond to resonant Raman scattering on optical phonons through free exciton state.

In real QW structures the free exciton motion is restricted by processes of exciton localization at disorder potential fluctuations (mainly fluctuation of concentration and fluctuation of QW width). Excitons with kinetic energies exceeding the amplitude of the potential fluctuations are extended states and can be considered as free particles with “good” k_{xy} wave vectors in the QW(xy) plane. Evidently it are these extended exciton states that are mainly responsible for the ω_1 line.

Figure 3 shows that the resonance profiles of the $\omega_2 - \omega_5$ components are in the limits of experimental error coincident with the distribution of emission intensity in the PL spectrum. It is well known that at low temperature photoluminescence correspond to emission of localized excitons [8]. Coincidence of the resonance profiles of the $\omega_2 - \omega_5$ lines with the shape of PL band indicates that the formation mechanism of the $\omega_2 - \omega_5$ REP components is similar to mechanism of localized exciton luminescence formation. Such coincidence is evidently connected with the fact that the shape of both PL band and the resonance profile is determined basically by the energy spectrum of density of localized exciton states which are distributed mainly in the region of the “virtual” exciton band bottoms.

The phonon spectrum of our ZnCdSe/ZnSe structure can be changed in respect to bulk ZnSe values due to several causes: phonon confinement, composition and strain effects. In contrast to previous works we have estimated the influence of all these causes on the phonon energies in our structures. Our estimates shown that it is possible to neglect the

influence of confinement at discussion of phonon spectra in our structures. At the same time the influence of composition and strain is rather noticeable. In a mixed ZnCdSe crystal two additional phonon modes are appeared in the restrahl band region in comparison with pure initial crystals [3]. Due to the difference in lattice constants both ZnCdSe and ZnSe thin layers in our structures are lattice-matched to the GaAs substrate. As a result both layers are biaxially strained by the substrate. The biaxial strain leads to shift and splitting of the energy of optical phonons [4]. As a result the structure of phonon modes with k_{xy} wave vectors in a strained ZnCdSe layer are determined by six characteristic optical phonon energies in the centre of the Brillouin zone, all these energies are being in the restrahl band region.

Analysis and comparison of the estimated characteristic phonon energies with energies of the $\omega_2 - \omega_5$ components in the hot luminescence spectra shown that they can be connected with interaction of localized excitons with phonons which are characteristic for strained ZnCdSe solid solution and wave vectors of those not far from the centre of the Brillouin zone ($k \sim 1/a_B$, where a_B is the exciton Bohr radius). Nevertheless it is not possible to exclude some influences of density of phonon states on the shape of REP spectra [1].

The energy of the Raman (ω_1) component in our spectra is rather close to and between the estimated values of LO values both in ZnCdSe and ZnSe strained layers. In our structure the thickness of ZnSe barrier regions is much larger than the thickness of ZnCdSe well. As a result, Raman scattering in the ZnSe regions could give some contribution to the observed spectra. However, scattering on ZnSe phonons hardly gives any input in the ω_1 line formation because difference between the energy of the observed ω_1 Raman component and the estimated energies of ZnSe LO phonons is larger than our experimental accuracy. Moreover, the intensity of the ω_1 line resonantly increases by more than two orders in the energy region of ZnCdSe exciton states. This resonance enhancement evidences that the Raman process is connected with ZnCdSe QW slab.

The ω_1 Raman process goes through k_{xy} extended exciton states. Due to wave vector conservation law the ω_1 phonon also must correspond to k_{xy} component. However the difference between the ω_1 energy and the energy of the k_{xy} component is larger than our experimental accuracy. At the same time the ω_1 energy is close to position which is characteristic for interface optical phonon. If our estimations are correct it can mean that Raman scattering goes through interface optical phonons.

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